Thermal fluctuations and anomalous elasticity of homogeneous nematic elastomers

Xiangjun Xing and Leo Radzihovsky
Department of Physics, University of Colorado, Boulder, CO 80309
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We present a unified formulation of a rotationally invariant *nonlinear* elasticity for a variety of spontaneously anisotropic phases, and use it to study thermal fluctuations in nematic elastomers and spontaneously anisotropic gels. We find that in a thermodynamic limit homogeneous nematic elastomers are universally incompressible, are characterized by a universal ratio of shear moduli, and exhibit an anomalous elasticity controlled by a nontrivial low temperature fixed point perturbative in $D = 3 - \epsilon$ dimensions. In three dimensions, we make predictions that are asymptotically exact.

Qualitatively important effects of fluctuations are generically confined to the vicinity of isolated critical points, where a system is tuned to be "soft", characterized by low energy excitations. As long as the ordered state is stable, fluctuations about it are typically described by a harmonic theory, controlled by a Gaussian fixed point. However, there exists a novel class of systems, that includes smectic[1, 2] and columnar liquid crystals[3], vortex lattices in putative magnetic superconductors[4], polymerized membranes[5], and nematic elastomers[6, 7, 8, 9], whose ordered states are a striking exception to this rule. A unifying feature of these phases is their underlying, spontaneously broken rotational invariance, that strictly enforces a particular "softness" of the corresponding Goldstone mode Hamiltonian. As a consequence, the usually small nonlinear elastic terms are in fact comparable to harmonic ones, and therefore must be taken into account. Similarly to their effects near continuous phase transitions, but extending throughout an ordered phase, fluctuations drive nonlinearities to qualitatively modify such soft states. The resulting strongly interacting ordered states at long scales exhibit rich phenomenology such as a universal nonlocal elasticity, a strictly nonlinear response to an arbitrarily weak perturbation and a universal ratio of wavevector-dependent singular elastic moduli, all controlled by nontrivial infrared stable fixed points illustrated in Fig.1.

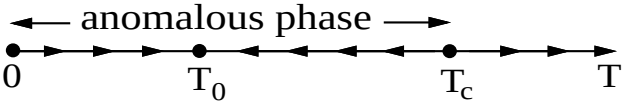

FIG. 1: Renormalization group flow for anomalously elastic solids, with T_c the transition temperature to the ordered state and T_o a nontrivial infrared stable fixed point controlling properties of the strongly interacting ordered, critical state.

Nematic elastomers and gels are weakly crosslinked polymer-liquid crystal composite materials, which, when nematically ordered exhibit fascinating elastic and electro-optic properties, and are therefore of considerable technological importance. The most unusual of these is the divergent strain response to a stress applied transversely to the spontaneous nematic direction[10, 11], an elastic softness that is a consequence of the novel nematoelastic Goldstone mode corresponding to a spontaneous orientational ordering and an accompanying elastic distortion of the network[6, 8, 12]. Despite of considerable progress in understanding the properties of these liquid crystalline rubbers[7], it was only recently appreciated, that (because of their harmonic softness) nematic elastomers and gels belong to the aforementioned class of anomalously elastic materials, and are the first example of three-dimensional solids with such novel fluctuation-driven properties.

In this Letter we present a unified formalism for a systematic derivation of the rotationally-invariant nonlinear elastic energy for variety of soft elastic media. This is a nontrivial ingredient that is a prerequisite to treating effects of fluctuations and external perturbations in such soft materials. We utilize this formalism to study long-scale elastic properties of homogeneous fluctuating nematic elastomers, and demonstrate that they indeed exhibit anomalous elasticity phenomenology discussed above[13].

A configuration of an elastic media is described by a mapping $\mathbf{x} \mapsto \vec{R}(\mathbf{x})$ from the D-dimensional reference space $\mathbf{x}$, which labels mass points of the media, to the d-dimensional target space $\vec{R}$. Our goal is to construct a *nonlinear* elastic energy functional $\mathcal{H}$, describing long-scale phonon deformations $\vec{u}(\mathbf{x}) = \vec{R}(\mathbf{x}) - \vec{R}^0(\mathbf{x})$ about the spontaneously ordered state $\vec{R}^0(\mathbf{x})$, with $\mathcal{H}$ exhibiting all rotational and translational symmetries of the reference and target spaces [8]. This is nontrivial to implement at a nonlinear level as some of the symmetries are spontaneously broken by the ground state, and are therefore not manifest (hidden) in $\mathcal{H}$.

A systematic way of constructing such $\mathcal{H}$ is by expanding it in in terms of *scalar* nonlinear operators S_n

$$\mathcal{H}[\underline{\underline{g}}] = \mathcal{H}[\underline{\underline{g}}_0] + a_n S_n + \frac{1}{2} a_{nm} S_n S_m + \dots \quad (1)$$

$$S_n = \mathrm{Tr}(\underline{\underline{g}}^n - \underline{\underline{g}}^n_0)\,. \quad n = 1, 2, \dots, \mathrm{Min(d,D)}, \quad (2)$$

with $g_{ij} = \partial_i \vec{R} \cdot \partial_j \vec{R}$ and $g^0_{ij} = \partial_i \vec{R}^0 \cdot \partial_j \vec{R}^0$ metric tensors corresponding to $\vec{R}(\mathbf{x})$ and $\vec{R}^0(\mathbf{x})$ configurations and summation convention over repeated indices is used

throughout. Eliminating the metric tensor

$$g_{ij} = g^0_{ij} + 2\frac{\partial R^0_\alpha}{\partial x_i} u_{\alpha\beta} \frac{\partial R^0_\beta}{\partial x_j} \tag{3}$$

in favor of the Lagrangian strain tensor

$$\begin{aligned} u_{\alpha\beta} &= \frac{1}{2}\left(\frac{\partial \vec{R}}{\partial R^0_\alpha}\cdot\frac{\partial \vec{R}}{\partial R^0_\beta} - \delta_{\alpha\beta}\right), &(4)\\ &= \frac{1}{2}(\partial_\alpha u_\beta + \partial_\beta u_\alpha + \partial_\alpha \vec{u}\cdot\partial_\beta\vec{u}) &(5)\end{aligned}$$

measured in terms of the ground state coordinate system R^0_α (rather than x_i) leads to the desired nonlinear elastic energy $\mathcal{H}[u_{\alpha\beta}]$. The necessity of this procedure is that it ensures that the expansion in $u_{\alpha\beta}$ about the spontaneously symmetry-broken ground state is rotationally invariant in both the target and reference spaces.

We have used this general formalism, with differences encoded in D, d and the nature of the broken ground state, $\vec{R}^0(\mathbf{x})$, to derive nonlinear elastic Hamiltonians for a variety of elastic media[14], some of which have previously appeared in the literature. In this Letter we apply it to study fluctuations in the most nontrivial and heretofore unexplored system, the spontaneously-uniaxial nematic elastomers with $d = D$[7, 8].

As discussed in detail in Refs. 6, 8, most of the properties of nematic elastomers can be captured by a purely elastic description in terms an elastic strain tensor, with a nematic order parameter $Q_{\alpha\beta} = S(\hat{n}_\alpha\hat{n}_\beta - \delta_{\alpha\beta}/3)$ (S and $\hat{n}$, respectively, the magnitude and the unit director for nematic order) integrated out. The isotropic-nematic transition is then characterized by a spontaneous uniaxial distortion $u^0_{\alpha\beta} \propto Q_{\alpha\beta}$, corresponding to a ground-state conformation and a metric tensor given by

$$\begin{aligned} \vec{R}^0(\mathbf{x}) &= \zeta_\perp \mathbf{x}_\perp + \zeta_z z\hat{z}, &(6)\\ g^0_{\alpha\beta} &= \zeta^2_\alpha \delta_{\alpha\beta}, &(7)\end{aligned}$$

with $\mathbf{x}_\perp$ a $D-1$-dimensional vector transverse to the nematic director $\hat{n} = \hat{z}$.

In the physical case of $D = d = 3$ there are three nonlinear rotationally invariant operators S_n, which, using Eqs.2 and 3, can be readily shown to be given by

$$\begin{aligned} S_1 &= 2\zeta^2_\alpha u_{\alpha\alpha}, &(8)\\ S_2 &= 4\zeta^4_\alpha u_{\alpha\alpha} + 4\zeta^2_\alpha\zeta^2_\beta u_{\alpha\beta}u_{\beta\alpha}, &(9)\\ S_3 &= 6\zeta^6_\alpha u_{\alpha\alpha} + 12\zeta^4_\alpha\zeta^2_\beta u_{\alpha\beta}u_{\beta\alpha} + 8\zeta^2_\alpha\zeta^2_\beta\zeta^2_\gamma u_{\alpha\beta}u_{\beta\gamma}u_{\gamma\alpha}, &(10)\end{aligned}$$

Using these expressions inside the expansion for $\mathcal{H}$, Eq.1, taken to quadratic order in S_n's, we obtain

$$\begin{aligned} \mathcal{H} &= a_z u_{zz} + a_\perp u_{ii} &(11)\\ &+ \frac{1}{2}\left(\mu_{zi}u^2_{zi} + B_z u^2_{zz} + \lambda_{zi}u_{zz}u_{ii} + \lambda u_{ii}u_{jj} + 2\mu u_{ij}u_{ij}\right)\\ &+ b_1 u_{zz}u^2_{iz} + b_2 u_{kk}u^2_{iz} + b_3 u_{ij}u_{iz}u_{jz} + c u^2_{iz}u^2_{iz},\end{aligned}$$

where Roman indices $(i, j, k, \ldots)$ take values in the $\perp$-space only and

$$\begin{aligned} a_{z,\perp} &= 2a_1\zeta^2_{z,\perp} + 4a_2\zeta^4_{z,\perp} + 6a_3\zeta^6_{z,\perp}, &(12)\\ \mu_{zi} &= 16a_2\zeta^2_z\zeta^2_\perp + 24a_3(\zeta^2_\perp + \zeta^2_z)\zeta^2_z\zeta^2_\perp,\\ &= \frac{4}{\zeta^2_z - \zeta^2_\perp}(\zeta^2_\perp a_z - \zeta^2_z a_\perp), &(13)\end{aligned}$$

with the detailed form of other elastic constants not important here[14].

In the absence of fluctuations, our expansion about the ground state $\underline{\underline{g}}^0$, characterized by $\zeta_{z,\perp}$ ensures that $a_z = a_\perp = 0$ in equilibrium. Given that in the nematic ground state, $\zeta_z \neq \zeta_\perp$, this then leads to a strict vanishing of the shear modulus $\mu_{zi} = 0$, as anticipated by Golubovic and Lubensky[6, 8] and is implicit in the neoclassical theory of nematic elastomers[7, 11]. It corresponds to a vanishing energy cost of a shear distortion u_{zi} displayed in Fig. 2*a*, that is one of many fascinating properties of nematic elastomers and is a manifestation of the "soft" Goldstone mode elasticity, discussed in the introduction. At finite temperature, fluctuations renormalize all elastic constants and in particular generate nonzero a_z, $a_\perp$. However, Ward identities associated with the underlying rotational invariance of $\mathcal{H}$ ensure that such linear in strain terms can always be eliminated by shifting to a true, thermally renormalized spontaneous deformation $\underline{\underline{g}}^0_R$[14].

Keeping only the most relevant terms in $\mathcal{H}$, Eq.12, and using nontrivial relations between elastic coefficients[14], enforced by the rotational invariance, we arrive at the effective nonlinear elastic Hamiltonian for a uniaxial nematic elastomer:

$$\begin{aligned} \mathcal{H}_{NE} &= \frac{B}{2}(w_{zz} + w_{ii})^2 + C(w_{zz} + w_{ii})(w_{zz} - w_{ii})\\ &+ \frac{\mu_L}{2}(w_{zz} - w_{ii})^2 + \mu\tilde{w}_{ij}\tilde{w}_{ij} + \frac{K}{2}(\nabla^2_\perp u_z)^2, &(14)\end{aligned}$$

where

$$\begin{aligned} w_{zz} &= \partial_z u_z + \frac{1}{2}(\nabla_\perp u_z)^2,\\ w_{ij} &= \frac{1}{2}(\partial_i u_j + \partial_j u_i) - \frac{1}{2}\partial_i u_z \partial_j u_z,\\ \tilde{w}_{ij} &= w_{ij} - \frac{w_{kk}}{D-1}\delta_{ij} &(15)\end{aligned}$$

are rotationally-invariant smectic-like and columnar-like nonlinear strain tensors relative to the uniaxial state,

$$\begin{aligned} B &= \frac{1}{4}(B_z + B_\perp + \lambda_{zi}), &(16)\\ \mu_L &= \frac{1}{4}(B_z + B_\perp - \lambda_{zi}), &(17)\\ C &= \frac{1}{4}(B_z - B_\perp), &(18)\end{aligned}$$

and $B_\perp = \lambda + 2\mu/(D-1)$ is the $\perp$-plane bulk modulus. It is easy to see that B is the overall bulk modulus, μ_L

and μ are the longitudinal and transverse shear moduli, respectively corresponding to deformations displayed in Fig. 2 b,c, and C couples bulk mode with longitudinal shear. Symmetry requires that transverse shear mode remains decoupled from other modes. Because of the vanishing μ_{zi}, we have also added a curvature K elastic term for u_z to ensure stability at long scales.

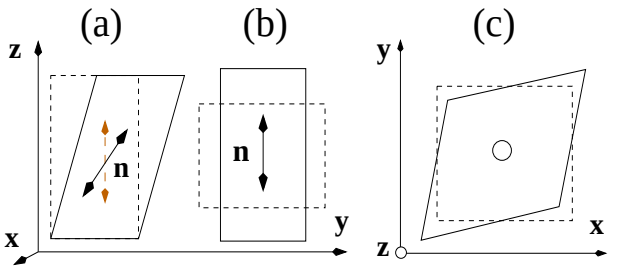

FIG. 2: (a) simple, (b) longitudinal and (c) transverse shear; in (c), nematic order is out of the paper. Simple shear transforms the system into an equivalent nematic ground state, and thus costs no energy.

Using $\mathcal{H}_{NE}$, we now proceed to study effects of thermal fluctuations on nearly homogeneous nematic elastomers. Standard analysis leads to the Fourier transform of harmonic correlators $G^0_{\alpha\beta}(\mathbf{x}) = \langle u_\alpha(\mathbf{x}) u_\beta(0)\rangle_0$:

$$G^0_{zz}(\mathbf{q}) = \frac{1}{B_z(1-\rho)q_z^2 + Kq_\perp^4}, \tag{19}$$

$$G^0_{zi}(\mathbf{q}) = -\left(\frac{\rho B_z}{2\mu+\lambda}\right)^{\frac{1}{2}} \frac{q_z q_i}{q_\perp^2(B_z(1-\rho)q_z^2 + Kq_\perp^4)}, \tag{20}$$

$$G^0_{ij}(\mathbf{q}) = \frac{B_z q_z^2 + Kq_\perp^4}{(B_z(1-\rho)q_z^2 + Kq_\perp^4)} \frac{q_i q_j}{(2\mu+\lambda)q_\perp^4} + \frac{1}{\mu q_\perp^4}(\delta_{ij}q_\perp^2 - q_i q_j), \tag{21}$$

where $\rho = (B-\mu_L)^2/(B_z(2\mu+\lambda))$.

To assess the role of nonlinear elastic terms in the presence of thermal fluctuations we compute perturbative corrections to elastic constants in $\mathcal{H}_{NE}$ and find that they are dominated by u_z fluctuations. Given the structure of G^0_{zz} correlator, as in smectic liquid crystals this perturbation theory diverges for $D \leq 3$ on length scales longer than $\xi^\perp_{NL} = \Lambda^{-1} g_\perp^{-1/(3-D)}$ $(= \Lambda^{-1} e^{32/(59 g_\perp)}$, for $D=3)$, with $g_\perp$ defined in Eq.24, below, and Λ an ultraviolet cutoff associated with the elastomer mesh size.

To understand the elastomer properties beyond ξ_{NL}, we perform a momentum-shell renormalization group calculation, perturbatively in $\epsilon = 3-D$. Integrating out short scale phonon fluctuations u_α and rescaling the spatial coordinates and fields, so as to keep Λ fixed, we arrive at the following flow equations for the elastic constants at scale $\Lambda^{-1}e^\ell$:

$$\begin{aligned}
\frac{dB}{d\ell} &= (D+3-3\omega-\eta_B)B,\\
\frac{dC}{d\ell} &= (D+3-3\omega-\eta_C)C,\\
\frac{d\mu_L}{d\ell} &= (D+3-3\omega-\eta_L)\mu_L,\\
\frac{d\mu}{d\ell} &= (D+3-3\omega-\eta_\perp)\mu,\\
\frac{dK}{d\ell} &= (D-1-\omega+\eta_K)K,
\end{aligned} \tag{22}$$

where,

$$\eta_B = \rho_2^2 g_L, \quad \eta_C = \eta_L = g_L, \quad \eta_\perp = \frac{1}{8} g_\perp, \tag{23}$$

$$\eta_K = \frac{g_L g_\perp \left(9 + 17\rho_1 - 22\sqrt{\rho_1}\rho_2\right) - 4{g_L}^2\left(-1+{\rho_2}^2\right)}{8\left(g_\perp \rho_1 + g_L\left(1+\rho_1 - 2\sqrt{\rho_1}\,\rho_2\right)\right)}.$$

The physics is controlled by the flows of two dimensionless coupling, $g_L(\ell)$ and $g_\perp(\ell)$, and two dimensionless ratios $\rho_1(\ell)$ and $\rho_2(\ell)$:

$$g_\perp = \frac{\mu}{4\pi K^{\frac{3}{2}}}\sqrt{\frac{(B-2\,C+\mu+\mu_L)}{B\,\mu+2\,C\,(-2\,C+\mu)+(4\,B+\mu)\,\mu_L}},$$

$$g_L = \frac{\mu_L}{\mu} g_\perp, \quad \rho_1 = \frac{\mu_L}{B}, \quad \rho_2 = \frac{C}{\sqrt{B\mu_L}}. \tag{24}$$

Flow equations for $\rho_1(\ell)$ and $\rho_2(\ell)$ are given by:

$$\frac{d\rho_1}{d\ell} = -g_L\,\rho_1\,(1-\rho_2^2), \tag{25}$$

$$\frac{d\rho_2}{d\ell} = -\frac{1}{2} g_L\,\rho_2\,(1-\rho_2^2), \tag{26}$$

where mechanical stability requires $|\rho_2| < 1$. With the exception of an unstable fixed point at $g_L^* = 0$ and an unstable fixed line at $|\rho_2| = 1$ (parameterized by ρ_1)[14], we expect (and verify a posteriori) that the infrared stable fixed point is characterized by $g_L^* \neq 0$. Equations 25,26 then imply that at such fixed point, $\rho_1^* = \rho_2^* = 0$. The flow equations for $g_L(\ell)$ and $g_\perp(\ell)$, the full form of which is too involved to be reproduced here[14], then simplify considerably:

$$\frac{dg_L}{d\ell} = \epsilon g_L - \frac{g_L\left(40{g_L}^2 + 68 g_L g_\perp + 13{g_\perp}^2\right)}{8\,(4g_L + g_\perp)}, \tag{27}$$

$$\frac{dg_\perp}{d\ell} = \epsilon g_\perp - \frac{g_\perp\left(4{g_L}^2 + 32 g_L g_\perp + 7{g_\perp}^2\right)}{4\,(4g_L + g_\perp)}. \tag{28}$$

In addition to reproducing the $3-\epsilon$-dimensional thermal smectic fixed point[1], with $g_\perp^* = 0, g_L^* = 4\epsilon/5$, we find an infrared stable fixed point at $g_\perp^* = 32\epsilon/59, g_L^* = 4\epsilon/59$, with

$$\eta_B = 0, \quad \eta_K = 38\epsilon/59, \tag{29}$$

$$\eta_C = \eta_L = \eta_L = \eta_\perp = 4\epsilon/59, \tag{30}$$

characterizing long-scale anomalous elasticity of homogeneous nematic elastomers[14].

For the physically most interesting case of $D = 3$, $g_L(\ell)$ and $g_\perp(\ell)$ are marginally irrelevant, allowing an asymptotically *exact* computation of anomalous elasticity. Standard asymptotic analysis of Eqs.25-28 with $\epsilon = 0$, shows that for large ℓ coupling constants flow to zero algebraically:

$$\rho_1(\ell) \sim \ell^{-4/59}, \quad (31)$$

$$\rho_2(\ell) \sim \ell^{-2/59}, \quad (32)$$

$$g_L(\ell) \approx \frac{1}{8} g_\perp(\ell) \approx \frac{4}{59\ell}. \quad (33)$$

Matching calculations, together with Eqs.22 then predict that on long scales all effective elastic constants except B are anomalous, depending logarithmically on wavevector $\mathbf{q}$ of the deformation

$$B(\mathbf{q}) \cong B_0, \quad (34)$$

$$\mu_L(\mathbf{q}) \sim \mu(\mathbf{q}) \sim C(\mathbf{q}) \propto (\log|\mathbf{q}_\perp|)^{-4/59}, \quad (35)$$

$$K(\mathbf{q}) \propto (\log|\mathbf{q}_\perp|)^{38/59}. \quad (36)$$

Hence at long scales, the bulk modulus is much larger than shear moduli, predicting that independent of microscopic details nematic elastomer is effectively incompressible in the thermodynamic limit. Logarithmically divergent strain-stress response immediately follows. Equations 24, 33 predict a universal ratio of the renormalized shear moduli

$$\frac{\mu^R}{\mu_L^R} = 8. \quad (37)$$

Although detecting logarithmic length scale dependence predicted here is likely to be difficult, the incompressibility of nematic elastomer and above universal ratio should be readily observable.

Based on our predictions in $3-\epsilon$ dimensions, we would expect a much stronger, power-law anomalous elasticity in two-dimensional (2D) nematic elastomers, with anomalous exponents given by Eq.29,30 with $\epsilon = 1$. However, a moment of reflection shows that because in 2D the subspace perpendicular to the nematic direction is one-dimensional, there is no transverse shear mode. We must therefore set $\mu = 0$, or deduce the properties 2D elastomers from a $D-1$-axial analytical continuation, as we have recently done for nematic elastomer membranes[15]. A more serious difficulty is that in our analysis we have ignored the subdominant columnar-like elastic nonlinearities, that, although irrelevant near $D = 3$, are relevant for $D < 5/2$ and in two dimensions are in fact identical to the smectic-like nonlinearities that we have studied here. Simultaneously treating both nonlinearities remains an interesting and challenging problem.

Throughout this work, we have also ignored elastomer heterogeneities. However, real elastomers are only *statistically* homogeneous and isotropic, and therefore results presented here are only valid up to a long but finite disorder-dependent length scale ξ_{NL}^{Δ}. On longer scales network heterogeneity dominates over thermal fluctuations and results presented here crossover to a distinct phenomenology, controlled by a nontrivial zero-temperature fixed point[2, 3, 4, 9].

To summarize, we have presented a unified formulation of a rotationally invariant nonlinear elasticity for a variety of elastic media with spontaneously broken spatial symmetry. We applied this formalism to nematic elastomers and studied their thermal fluctuations. We found that they exhibit anomalous elasticity, which in three dimensions is characterized by a logarithmic wavevector dependence of elastic moduli, universal ratio of shear moduli, and a long-scale incompressibility.

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[13] In a recent work O. Stenull and T. C. Lubensky found results that are in agreement with those presented here.

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